MILD GASIFICATION OF COAL IN A MOVING BED REACTOR

Muthu S. Sundaram*, Peter T. Fallon and Meyer Steinberg
Process Sciences Division
Brookhaven National Laboratory
Upton, New York 11973, U.S.A.

ABSTRACT

The Brookhaven National Laboratory (BNL) tubular moving-bed reactor was used to perform gasification experiments with Wellmore Kentucky No. 8 bituminous coal under mild reaction conditions at two levels of the process variables: temperatures of 550° and 650°C, coal particles residence times of 0.1 min. and 2 min.; and lime addition of 0% and 10% of the coal fed, with inert N_2 as the sweep gas. Using a fractional factorial statistical design, the interactive effects of process variables were evaluated. The highest total hydrocarbon yield of 22.6% (MAF) was obtained at 650°C and 2 min. coal residence time in the presence of 10% lime additive. The gaseous hydrocarbon yield added only another 0.8% to the total hydrocarbon yield under the above conditions.

The effect of coal residence time on total hydrocarbon yield was significant only at the higher temperature level of 650°C and in the presence of the lime additive. In general, increase in residence time, other conditions being equal, tends to decrease the retention of sulfur in char. The presence of lime in the feed had a beneficial effect on the retention of sulfur in the char only at low residence times.

INTRODUCTION

A large body of literature exists on the effects of process variables on product yields from pyrolysis and gasification of coal at high temperatures, often in the range of 900° to 1000°C. However, the independent and interactive influence of factors affecting the mild gasification behavior of coals has not been studied in a systematic manner. Mild gasification is defined as devolatilization at temperatures up to 700°C, and at low pressures, less than 50 psi. The idea is to produce condensible hydrocarbon fuel from coal in a low cost process under mild reaction conditions and use the char for boiler fuel. The effects of temperature, coal particle residence time and lime additive on the yields of products from the mild gasification of Wellmore Kentucky No. 8 bituminous coal are presented herein.

EXPERIMENTAL

Figure 1 shows a schematic of the experimental reactor which consists of two sections. The top section ia a 1-in. I.D. x 11-ft long entrained tubular reactor and the bottom is a 4-in. I.D. x 6-in. long moving bed reactor. The top section can be used under two separate sets of conditions; (1) to obtain short coal particle residence times in the order of a few seconds and (2) to preheat the coal particles to various temperatures for feeding into the moving

bed reactor. A motor-driven vaned agitator keeps the coal moving through the moving bed reactor and empties the char into an unheated collection vessel below. The product gas lines are located between the reactor and liquid condensers in a manner so as to prevent tar accumulation in the ports and the transfer lines. A liquid vythene scrubber is used to scrub the gases to collect any trace hydrocarbon carryover.

An on-line process gas chromatograph is used to measure all the light gases and hydrocarbon products. The condensible liquid products and the char are collected separately. From the elemental analyses of all the products, a complete mass balance is obtained.

The reported experimental yields of coproducts include five categories of hydrocarbons: (1) benzene-toluene-xylene (BTX), (2) hydrocarbon (HC) liquids which consist of all HC liquids heavier than BTX including oxygenates and tars, (3) total liquid HC condensibles, which is the sum of BTX and HC liquids, (4) gaseous HCs mainly CH_4, C_2H_6, and C_2H_4 (CO and CO_2 are analyzed and reported separately, and H_2 and H_2O are determined by difference), and (5) total HCs which is the sum of all liquid and gaseous hydrocarbons. The char collected consists of all the solid non-volatile residue containing no benzene extractable liquids.

<center>RESULTS AND DISCUSSION</center>

A total of eight experimental runs were performed using Wellmore Kentucky No. 8 bituminous coal. The experimental conditions and the yields of the products are shown in Table 1.

In a conventional approach, the value of one process variable is changed at a time, thereby estimating its influence at constant conditions for other variables. This, however, assumes that there is no interaction between the variables. This has a potential of leading to erroneous conclusions. The data in Table 1, on the other hand, provides information on the interactive effects of temperature, residence time and lime addition on the product yields in which more than one variable is changed at a time. In other words, the influence of all variables are estimated throughout the range of each and every variable.

The experimental yields were correlated in accordance with a fractional factorial statistical method. The data from Table 1 can be clearly shown in a three-dimensional box representation in which the interactive effect of all three variables can be represented simultaneously along the three axes of the cube. This is superior to the conventional bivariate plots.

Figure 2 shows the yield of total hydrocarbon products as a function of temperature, residence time and percent lime addition. The circled data are the experimental values at a given condition rounded off to the nearest whole number and the number in parenthesis represents the corners of the cube. The numerical differences in yields at two different conditions are also shown in the figure along the axes with an appropriate sign.

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The highest total hydrocarbon products yield was 23.4% (MAF), and it occurred at 650°C in the presence of 10% lime at a 2 min. residence time. At a given residence time, the incremental yield of total hydrocarbon products remained virtually the same at all levels of lime addition tested so far. For example, at all residence times, the total hydrocarbon yield increased by 17 percentage points as a result of an increase in temperature from 550° to 650°C, with 10% lime. Similar effects were noticed when no lime was added to the feed, however, the actual incremental yield is almost halved (17% vs. 9-10%) when compared with the data with 10% lime.

At a given temperature, the effect of residence time and lime addition on total hydrocarbon yield is complex. At 550°C, there is a decrease of 4 percentage points, from 0.1 min. to 2 min. residence in the absence of lime, whereas with 10% lime additive, there is no change in the yield at all. On the other hand, at 650°C, and with 10% lime, there is a marked increase in the total hydrocarbon yield with increase in residence time (8% points).

At higher residence times, there is a slight increase in the yield (3% points) when 10% lime is added (3% vs. 6% at 550°C and 20% vs. 23% at 650°C). At 0.1 min., under identical conditions, a small decrease is seen (7% vs. 6% at 550°C and 17% vs. 15% at 650°C), which corresponds to no change when experimental error is taken into consideration.

The estimates of the individual and interactive effects of the variables can be made via Yate's algorithm, the details of which are available [1,2]. The confounding pattern and Yate's algorithm for the production of total hydrocarbons is shown in Table 2. Figure 3 is a normal probability plot of the estimated effects in Table 2.

Even though the temperature seems to be apparently more effective than any other variable, it should be noted that the observed results can not be explained on the basis of temperature alone. An interactive effect involving the variables, temperature and lime addition, seems to be more significant. At a higher temperature, reactions requiring higher activation energies, which would not otherwise occur at a lower reaction temperature, become more probable. The removal of primary tar via secondary cracking reactions on the surface of the added lime to produce lighter hydrocarbons seems to be favored by an increase in residence time which explains a higher incremental yield of 8 percentage points from 15% (at 650°C, 0.1 min., 10% lime) to 23% (at 650°C, 2 min., 10% lime). This is in agreement with earlier reports on the catalytic effect of inorganic materials on the decomposition of primary tar [3].

The variation in total hydrocarbon condensible product yield with temperature, residence time and lime addition is shown in Figure 4. The effect of the process variables on the liquid hydrocarbon yield is similar to that on the total hydrocarbon (liquid and gas) yield. The yield of gaseous hydrocarbons is much smaller than the liquid hydrocarbon yield. No detectable hydrocarbon or other gases were produced at 550°C. The highest observed gas yield

was 1% at 650°C and 0.1 min. residence time. This indicates that the experimental conditions are much more favorable for the production of liquid hydrocarbons.

At 550°C, the BTX yield was constant at 1.6% MAF and was not affected by residence time or lime additive (Figure 5). At 650°C, the BTX yield varied between 3.1% and 4.4% depending on other conditions. At 650°C, addition of lime increased the BTX yield by 0.4 percentage points both at 0.1 min. and 2 min. residence times; at the same temperature, increase in residence time caused an increase in BTX yield of 0.9 percentage points, both with no lime and with 10% lime addition. From this, it can be inferred that the BTX yield is influenced by an interactive effect involving the variables, temperature and residence time.

From an elemental analysis of the hydrocarbon liquids, the hydrogen to carbon ratio was calculated. The H/C ratio varied between 0.85 to 1.08 (Figure 6). In general, at 550°C, the H/C ratio of the liquid was close to unity (average of 1.03) and at 650°C, the H/C was slightly less than unity (average of 0.93). At 650°C, either addition of lime or increasing the residence time, increased the H/C ratio of the liquid product.

The primary purpose of the addition of lime to the coal feed was to increase the retention of sulfur in the solid residue. The sulfur content of the char was determined via elemental analysis. From a knowledge of the sulfur material balance, the percentage of the sulfur in the feed coal that remained in the char was calculated. The results are pictorially represented in Figure 7 as before. The presence of lime in the feed had a significant effect on the retention of sulfur in the char only at low residence times. For example, at 0.1 min., addition of 10% lime to the feed increased sulfur retention in the char by 12 to 14 percentage points. In general, temperature has slight negative affects and an increase in residence time, other conditions being equal, tends to decrease the retention of sulfur in char. As much as 91% of the original sulfur was retained in the char at 550°C, 0.1 min. residence time and in the presence of lime. However, it should be remembered that the total hydrocarbon yield under these conditions was low at 6.4% (MAF). At conditions of maximum hydrocarbon production (22.6%), sulfur retention in the char was only 79%.

<center>CONCLUSIONS</center>

A summary of the conclusions for the effect of process variables on the Wellmore Kentucky No. 8 coal are as follows:

● The highest total condensible hydrocarbon yield of 22.6% (MAF) was obtained at 650°C and 2 min. coal residence time in the presence of 10% lime additive. The gaseous HC yield adds only another 0.8% to the total HC yield.

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- Even though the absolute yields were different, the trend in the total condensible hydrocarbon yield was similar at both residence times of 0.1 and 2 min. For example, at the lower residence time of 0.1 min., increasing the temperature from 550° to 650°C resulted in an 8 to 10 percentage points increase in the hydrocarbon condensible yield, both with and without the lime additive. At the higher residence time of 2 min., the increase in the yield was 16 percentage points, all other conditions remaining the same.

- The effect of coal residence time on total hydrocarbon yield was significant only at the higher (650°C) temperature level and in the presence of the lime additive. For example, at 650°C and in the presence of lime, there was an increase of 8 percentage points in absolute yield which amounts to about a 55% increase in the relative yield (14% MAF to 22% MAF).

- The presence of lime in the feed had a beneficial effect on the retention of sulfur in the char only at low residence times. In general, an increase in residence time, other conditions being equal, tends to decrease the retention of sulfur in char.

- As much as 91% of the original sulfur was retained in the char at 550°C, 0.1 min residence time and in the presence of lime; however, the total HC yield under these conditions was low at 6.4% (MAF). At conditions of maximum condensible hydrocarbon production (22.6%), sulfur retention in the char was only 79%.

- The H/C ratio of the total hydrocarbon condensibles is found to be close to unity (1.0). This indicates the highly aromatic composition of the condensibles.

REFERENCES

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2. Box, G. E. P., Hunter, W. G., and Hunter, J. S. Statistics for Experiments. An Introduction to Design, Data Analysis and Model Builders, John Wiley and Sons, New York, NY, 1978.

3. Franklin, H. D., Peters, W. A., and Howard, J. B. ACS Fuel Division Prepr., 26 37 (1981).

Table 1. Experimental Yields of Products from Mild Gasification of Coals.
Effect of Process Variables
N_2 Sweep Gas 15 psig

| Run No. | Temp. °C | Res. Time (min) | Additive % lime | BTX | Conversion, Wt% MAF Feed Coal | | | |
					HC liquids	Total HC Cond.	Gas	Total
1042	550	0.1	0	1.6	5.4	7.0	0.0	7.0
1047	650	0.1	0	3.1	13.9	17.0	0.2	17.2
1041	550	2.0	0	1.6	4.8	6.4	0.0	6.4
1043	650	2.0	0	4.0	10.2	14.2	0.9	15.1
1040	550	0.1	10	1.6	1.1	2.7	0.0	2.7
1045	650	0.1	10	3.5	15.3	18.8	1.0	19.8
1044	550	2.0	10	1.6	4.5	6.1	0.0	6.1
1027	650	2.0	10	4.4	18.2	22.6	0.8	23.4

Table 2. Yate's Algorithm for the Production of Total HC from Kentucky No. 8.

Run No.	Yield Wt % Coal Feed	(1)	(2)	(3)	Divisor	Estimate	Effect
1042	7.0	24.2	45.7	97.7	8	12.2	Average
1047	17.2	21.5	52.0	53.3	4	13.3	T
1041	6.4	22.5	18.9	4.3	4	1.1	R
1043	15.1	29.5	34.4	1.3	4	-0.3	TR
1040	2.7	10.2	-2.7	6.3	4	1.6	A
1045	19.8	8.7	7.0	15.5	4	3.9	TA
1044	6.1	17.1	-1.5	9.7	4	2.4	RA
1027	23.4	17.3	0.2	1.7	4	0.4	TRA

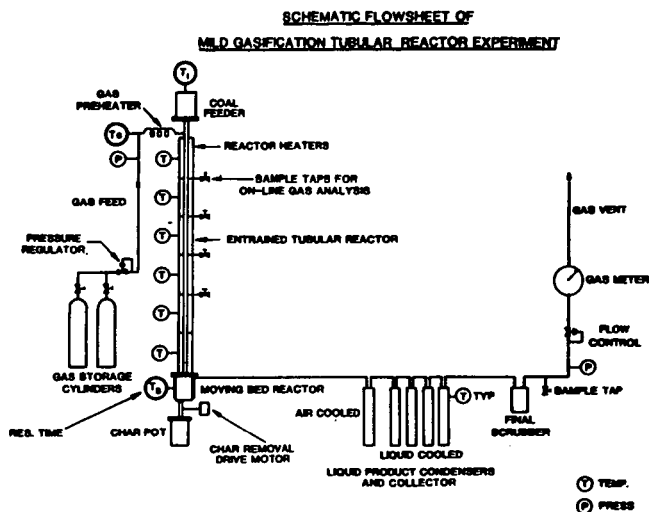T = temperature; R = residence time; A = % lime additive.

(1.9)

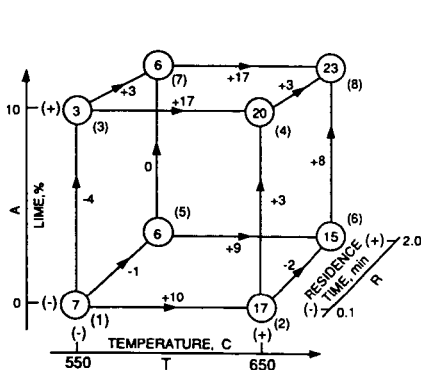Figure 1. Schematics of Experimental System.

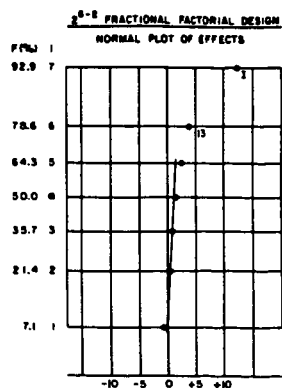Figure 2. Total Hydrocarbon Yield from Kentucky No. 8 Coal.

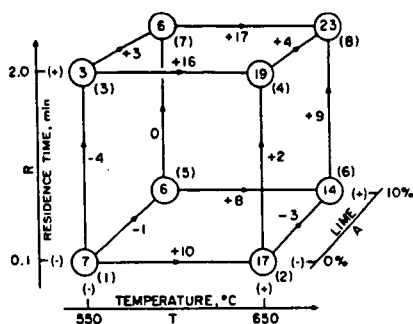Figure 3. Normal Probability Plot of Total Hydrocarbon Yield Contrasts from Kentucky No. 8 Coal.

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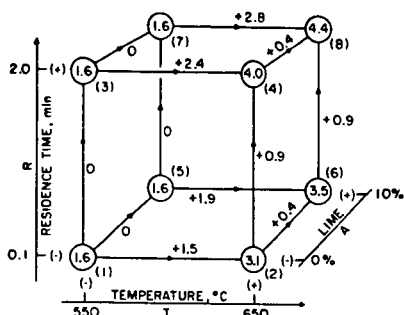Figure 4. Total Hydrocarbon Condensible Liquid Yield from Kentucky No. 8 Coal.
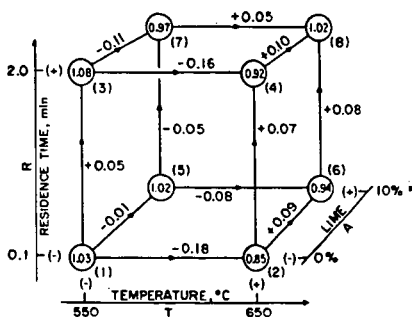

Figure 5. BTX Yield from Kentucky No. 8 Coal.

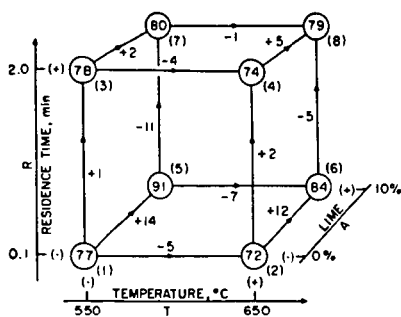Figure 6. H/C Ratio of Hydrocarbon Liquids from Kentucky No. 8 Coal.

Figure 7. Sulfur Retention in the Char of Kentucky No. 8 Coal.

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